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NOTE ON THE LINEAR FORCE OF GROWING  
CRYSTALS

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In 1905 we published a short paper,<sup>1</sup> qualitative in character, with the purpose of demonstrating from simple laboratory measurements the existence of a linear force, apart from the volume expansion, exerted by growing crystals. We sought to show (1) that when a crystal, fed with appropriate saturated solution, grows in an open crack between walls with which it comes into contact on both sides, pressure is exerted to separate the walls, notwithstanding unrestricted opportunity for growth in other directions; (2) that the linear force thus exerted is of the order of magnitude of the breaking strength of the crystal and therefore a geologic force of considerable magnitude and importance.

The crucial experiment offered in support of this conclusion was prepared in an ordinary crystallizing dish upon the bottom of which was cemented a block of plate glass having a plane upper surface. A well-formed crystal of alum was laid upon this plane surface, and upon it a second plane glass plate carrying a weight. A saturated solution of alum was poured into the crystallizing dish in sufficient quantity to cover the crystal, and then left to evaporate quietly under conditions reasonably free from temperature change and from

<sup>1</sup> "The Linear Force of Growing Crystals," *Proc. Wash. Acad. Sci.*, VII (1905), 283.  
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dust. The original drawing of the experimental arrangements is reproduced here for the sake of definiteness (Fig. 1). Saturated solution was added from time to time if needed, so that throughout the experiment the crystal remained submerged in its saturated solution. The thickness of the crystal was measured at intervals with an appropriate instrument.

The experiment was repeated many times with a single crystal of alum and various weights, and also with a crystal of copper sulphate, of potassium ferrocyanide, and of lead nitrate, in appro-

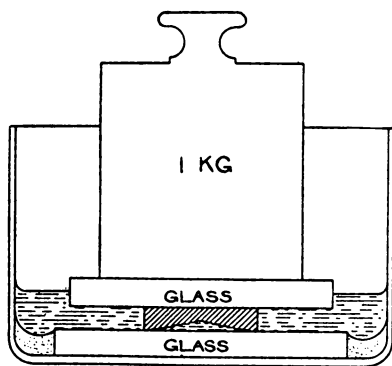


FIG. 1.—A crystal shown growing between two glass plates and lifting a heavy load. Reprinted from the paper of 1905 (*op. cit.*).

priate solutions. In no single instance during this series of observations did the crystal fail to lift (1) its own weight; (2) the weight of the superimposed glass plate; (3) the weight of the load upon the glass plate. The distance through which the load was lifted varied in different experiments from a few hundredths to 0.5 mm.<sup>1</sup>

In 1913 Bruhns and Mecklenburg<sup>2</sup> published a series of experiments upon the same subject which purported to repeat those which we had made and to disprove them. They announced their inability to obtain the experimental results which we have described, and categorically denied, in the face of much corroborative evidence contained in their own paper, the existence of such a linear force exerted during the growth of crystals.

To this paper we did not at once reply because it seemed impossible that other investigators would long allow the crucial experi-

<sup>1</sup> The data upon which this preliminary account is based were obtained during the winter of 1902-3, but the records were burned in the Geological Survey fire in the following year, which may serve to explain the absence of complete data in the publication of 1905.

<sup>2</sup> W. Bruhns and Werner Mecklenburg (Clausthal), "Über die sogenannte Kristallisationskraft," *Jahresbericht des Niedersächsischen geologischen Vereins zu Hannover*, VI (1913), 92.

ment in a question of fact to remain unverified where a principle of such far-reaching importance to geologists was at stake. The issue could have been put to the test by anyone in a few hours without special facilities of any kind.

When, however, a distinguished physical chemist (Boeke, in his admirable book, *Grundlage der physikalisch-chemischen Petrographie*, 1915, p. 328) accepts their conclusion without test, and casts out the "linear force of growing crystals" from geological calculation entirely, a form of protest appears necessary. This outcome is the more unfortunate because it must be patent to a physical chemist that Bruhns and Mecklenburg did not in fact repeat our simple experiment at all, but substituted one which is not, by itself, conclusive upon the point at issue; and, further, because all of their recorded experimental evidence is entirely in accord with our experience and conclusions. There appears to be no contradiction of fact, but only one of appropriate interpretation; the conditions which govern the behavior of a single loaded crystal are modified when an unloaded crystal is introduced into the same solution, as was done by Bruhns and Mecklenburg.

Now upon the main question of fact as to whether a crystal of alum or other substance will or will not lift a load when immersed in its saturated solution in an open vessel, nothing is simpler than to repeat the experiment which we described. An ordinary open crystallizing dish, an alum crystal placed on the bottom of it and covered with a saturated solution of the same liquid, and an ordinary brass weight of one or two hundred grams upon the crystal, together with a simple apparatus to be found in any laboratory for measuring the thickness of the crystal before and after the experiment, provide all the equipment necessary to establish or disestablish the fact of growth in the direction of the load. Two fairly typical cases follow:

These experiments are so straightforward, and withal so conclusive in their results, that it would hardly seem possible to go astray; nevertheless, Bruhns and Mecklenburg, in the paper above referred to, have denied their validity. A series of measurements taken from the paper of Bruhns and Mecklenburg is quoted in Table II.<sup>1</sup>

<sup>1</sup> *Op. cit.*, p. 100.

This is not a simple repetition of our experiment, although it has been made to appear so. Bruhns and Mecklenburg have placed in the same vessel a loaded crystal *and an unloaded crystal*, and have observed, as might have been anticipated, that the unloaded crystal increased in thickness while the loaded crystal did not. This result was confirmed in other measurements of the same kind which need not be reprinted here.

TABLE I\*

CONDITIONS AS ABOVE DESCRIBED; ROOM TEMPERATURE  
ABOUT 20°; LOAD 95 GM. NO OTHER CRYSTAL  
PRESENT

Time (Hours)	Thickness of Crystal	Increase of Thickness
0.....	8.3260 mm.	0 mm.
22.....	8.4400	0.114
98.....	8.4569	0.131

A VERY SMALL CRYSTAL, CONDITIONS AS BEFORE,  
LOAD 0.7 GM.

Time (Hours)	Thickness of Crystal	Increase of Thickness
0.....	3.7649 mm.	0 mm.
20.....	3.7803	0.015
50.....	3.8027	0.038
68.....	3.8029	0.038
145.....	3.8262	0.061

\*The experimental results contained in the present paper were courteously placed at our disposal by Mr. J. C. Hostetter, of the Geophysical Laboratory, who will report in greater detail upon this problem in the near future.

Let us consider for a moment the conditions of crystal growth in a saturated solution. Suppose a single isometric crystal to be immersed in a solution saturated with respect to it; and suppose further that the water is gradually removed from the solution by evaporation, thus inducing potential supersaturation and the continued growth of the crystal in consequence. If the supersaturation is greater than can be balanced by the growth of this crystal under the prevailing conditions, other nuclei will tend to form upon which deposition may take place. Now, what will happen when two crystals of the same substance are present, one of them being

less stable than the other owing to its inferior size, or because it is strained, or because it is an inherently instable form, or for any other reason? In the first place, it is plain that a solution to be in equilibrium with a less stable crystal will require to be more concentrated than that in equilibrium with the more stable one; consequently in the foregoing case the solution will become super-saturated with respect to the more stable crystal before it is super-saturated with respect to the other, and the former will begin to grow before the latter, which, indeed, will not grow at all (it may even dissolve) unless the degree of supersaturation is greater than the growth of the more stable crystal can keep balanced.

TABLE II

A LOADED (1 GM.) AND AN UNLOADED CRYSTAL IN THE SAME SOLUTION.  
TEMPERATURE = 10° C.

DATE	UNLOADED CRYSTAL		LOADED CRYSTAL	
	Thickness of Crystal	Increment of Thickness	Thickness of Crystal	Increment of Thickness
May 5, 1913.....	9.68 mm.	.....	10.00 mm.	.....
June 5, 1913.....	10.22	+0.54 mm.	10.06	+0.06 mm.
July 5, 1913.....	10.86	+0.64	10.04	-0.02
September 5, 1913.....	11.42	+0.56	10.06	+0.02

Analogous cases also arise when the crystals are of equal stability but the solution is inhomogeneous, and there results from the action of outside forces (imperfect stirring, thermal convection, gravitative adjustments) a distribution of concentration such that one crystal is in contact with solution of higher concentration and grows while the other cannot.

A familiar instance may be cited. If in an unstirred saturated solution in a closed vessel two identical crystals are placed, of which one is suspended a few millimeters above the other, the lower crystal will grow while the upper one dissolves slowly. And similarly under like conditions the bottom of a very large crystal will grow at the expense of the top, and the prismatic lateral faces gradually acquire the contour of a flight of steps; the reason in either case is that under the action of gravity the solution tends to become more concentrated in its lower layers, and hence, since it is kept

saturated at the level of the upper crystal, it will be potentially supersaturated at the level of the lower. In short, the rate of growth of an isometric crystal depends altogether upon whether the concentration of the layer of solution in contact with it is or is not potentially supersaturated with respect to that particular crystal. One serious result of this is that if diffusion toward a certain face is obstructed (e.g., when that face lies against a glass plate), that face will be unable to grow like the other faces. To this we shall revert.

Now it is the importance of this principle to the question under consideration which has been overlooked by Bruhns and Mecklenburg. As soon as it is appropriately applied, their observations correlate perfectly with ours.

The effect upon the saturation concentration of differently oriented faces of the same crystal in non-isometric systems is a matter less well understood and is beyond the scope of this inquiry. We may therefore omit further consideration of it in this connection.

In plain terms and without taking account of unnecessary complications, the situation in a saturated solution, under the conditions now under consideration, may be described somewhat in this way. Given a body of saturated solution of a salt in an open vessel, the amount of the dissolved substance which can remain in solution for a given temperature is limited, and it may begin to separate out either when the temperature is changed or when continued evaporation from the free surface of the liquid has sufficiently increased the concentration. Differences in the concentration due to the slowness of diffusion will cause gravitative readjustments, bringing the portions containing the maximum amount of dissolved matter to the bottom. A single crystal of the salt exposed in the bottom of the vessel will now grow upon its exposed faces, and if the rate of evaporation is not too great, this growth may take care of all of the excess of solute resulting from the evaporation process. If the evaporation proceeds at a greater rate, other nuclei will form, and if two or more crystals are feeding upon the product of the evaporation their relative rate of growth (+ or -) may depend upon relative stability, position, size, or the amount and distribution of load.

Inasmuch as a crystal lying upon the bottom of an evaporating dish must rest upon one of its faces, this face will support a load represented by the weight of the crystal less the buoyancy correction, and this load will impose limiting conditions upon the rate of growth upon this face when compared with the neighboring faces, as was pointed out in our paper in 1905, and by Bruhns and Mecklenburg in 1913. A weight placed upon the crystal merely adds something to the total load supported by this face (and perhaps covers an additional portion of the crystal surface), *but contributes no new factor to the problem*. If this superimposed weight is very large, the distribution of the resulting strain may become important, but these are questions of degree only. Limitations of circulation or diffusion in the capillary liquid layer below the crystal are affected by the amount and distribution of the aggregate load, and not at all by its character (whether crystal substance or foreign matter). This fact would hardly appear to require demonstration, but has certainly caused some confusion, nevertheless.

It is then clear that the exposed top and side faces (or the side faces alone if the top is covered) may grow freely while the bottom remains more or less undernourished, depending upon the load which is supported there and the consequent impairment of circulation. Nevertheless, if the degree of supersaturation and the amount of material which is being furnished to the crystal through evaporation and diffusion is sufficient in quantity and properly circulated, the saturation concentration opposite that face also, that is, in the thin layer of liquid upon which the crystal rests, may be reached and the crystal may grow upon the bottom as well as upon the sides. Failure of the circulation in this supporting layer may, and in fact usually will, restrict the growth here to the periphery of the supporting face, causing it eventually to rest upon a thin outer rim<sup>1</sup> of new growth rather than upon its initial flat surface, but growth will nevertheless take place here as elsewhere. The

<sup>1</sup> As was pointed out in our paper in 1905, these supporting rims are often so thin as to debar the usual methods of area measurement. At that time an approximate measurement was obtained by inking the crystal with an insoluble ink and printing its impression upon a plane glass plate coated with white celluloid. The impressions thus secured contain lines so fine as to defy reproduction by the usual means and probably yield but a rough approximation of the surface area which supports the load.



greater the crystal, the greater the weight supported upon its contact surface (or rim), and the greater, a fortiori, the difficulty of reaching the saturation concentration in any portion of the supporting liquid layer and providing for further growth from the bottom.<sup>1</sup>

To make specific application of this analysis to our case, namely, a single alum crystal resting upon a thin liquid layer in its own saturated solution and supporting an outside load, the most favorable surfaces for growth will be the lateral faces, and there the

<sup>1</sup> That the description of these phenomena as observed by Bruhns and Mecklenburg differs in no essential particular from our own may be seen from the following extracts from their paper (*op. cit.*):

P. 97: "The common view of crystal growth has hitherto been that a crystal grows exclusively through additions of new matter from without. A crystal can therefore grow only where there is surface in contact with the solution, which offers room for new material to be added, and where expansion can occur. A great deal of experience and many observations are in accord with this view."

P. 100: "Unloaded crystals show continuous growth and the increase in weight, which diminishes in the case of loaded crystals approximately in proportion to the decrease in exposed area, proceeds normally; that is, where new material can find a foothold there is growth, where it cannot there is none."

P. 102: "A further circumstance which requires to be considered in connection with the formation of the cup-shaped base [see Fig. 1] is this, that in a laboratory experiment in a glass vessel the base of the crystal is not in contact with the vessel but rests upon a layer of water or of solution which adheres both to the glass and to the crystal. The occurrence of such adhesion or adsorption layers is sufficiently familiar; they are lacking only when the crystal grows fast to its support. By reason of this liquid layer between the crystal and its support, the supersaturated solution is enabled to diffuse under the crystal, even though the rate of diffusion in the capillary layer is smaller than elsewhere. It will not advance far, however, for the molecules in excess of the quantity needed to saturate the solution, when they attempt to pass close under the crystal, will be quickly caught, that is, a rim [*Wulst*] will grow beneath the periphery of the crystal, as observation in fact shows."

P. 103: "Crystals which form on the bottom of the vessel, without exception show cup-shaped bases."

P. 103: "If a large alum crystal is laid upon a smooth surface in a saturated solution which is evaporating, its lower surface becomes cupped, but not in regular steps like rock salt or bismuth. Instead of this the central portion remains practically flat while a narrow supporting rim grows about it. Neither does long-continued growth result in a stepped formation; the narrow rim moves outward while the inclosed area continues nearly or quite flat."

P. 105: "The explanation of the phenomenon may perhaps be that the supporting rim—which must of course bear the weight of the crystal—possesses a higher solubility than the remainder of the crystal. Possibly also there is a difference in solubility in different crystallographic directions."

potential supersaturation will first be reached. If the rate of evaporation is sufficiently great, the saturation concentration in the liquid layer below the crystal will also be reached, in whole or in part, and growth from below will go on, albeit at an appropriately diminished rate compared with the sides because of the load and the deficiency of circulation. If the upper surface is covered by the load, it will share the limitation of circulation of the bottom surface. If the load upon the crystal is too great, or the rate of evaporation slow, the saturation pressure may not be reached anywhere in the supporting liquid layer, and growth here may be stopped. A still further increase in the load may even cause resolution of the bottom surface, while the side surfaces continue to grow.

In support of this analysis, the accompanying photograph (Fig. 2) of a crystal grown under a heavy load and the measurements made upon it (Table III) will be found interesting.

A single crystal of potash alum was immersed in a solution saturated with both potash and chrome alum under a load of 190 gm. Other conditions were as heretofore described. The dark areas

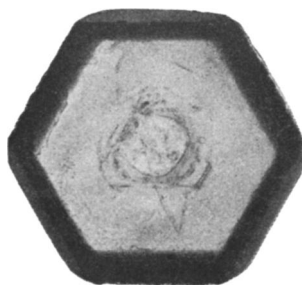


FIG. 2.—A crystal grown under load. Dark portions are new growth (under load). (View from above.)

are fresh deposit, colored of course by the chrome alum and thus distinguishable from the original crystal. It is plain that there is no

TABLE III  
CONDITIONS AS BEFORE. LOAD 190 GM.

Time (Hours)	Thickness of Crystal	Increase in Thickness
0.....	3.5090 mm.	0 mm.
17.....	3.5239	0.015
65.....	3.5859	0.077
113.....	3.5926	0.084

fresh deposit (colored matter) on the central portions of the original crystal above or below. The accessions of fresh matter are found exclusively on (and beneath) the new lateral faces. Nevertheless,

this crystal lifted its load (Table III), and we are concerned at the moment less with the form than with the fact of linear growth in the direction of the load. Bruhns and Mecklenburg have recognized and pictured this peripheral rim (*Wulst*) and admit its lifting action for increasing loads of crystal substance,<sup>1</sup> but deny lifting power when crystal substance is replaced by foreign substance.

The distribution of growth about an unloaded crystal is rather well shown by the same device. Fig. 3 is a vertical section through an unloaded crystal of potash alum grown in a solution saturated



FIG. 3.—Distribution of new growth about an unloaded crystal.

with both chrome and potash alum. The color plainly reveals the distribution of new crystal substance and shows the original crystal, together with the mass of new matter deposited upon its upper surface, to have been lifted bodily 0.4 mm. by the new growth. It may be

noted also that the original crystal was inverted from its position of original growth (its cup is still distinguishable facing upward) in order that subsequent growth might suffer no modification through special limitations of circulation imposed by the original supporting rim.

By the same reasoning, the case offered by Bruhns and Mecklenburg (Table II) is capable of equally definite analysis. Here we have in the same solution two crystals, one loaded and the other not. The saturation concentration will ordinarily be first reached upon the top and side surfaces of the unloaded crystal; secondly, in the exposed (and strained) side surfaces of the loaded crystal; thirdly, in the supporting liquid layer beneath the unloaded crystal; and last of all, in the liquid layer beneath the loaded crystal. Whether this last concentration can be reached in the presence of the three lower saturation concentrations, all of which are exacting their toll of the solution, will depend upon fortuitous relations of

<sup>1</sup> See Bruhns and Mecklenburg's paper, p. 105; also the quotations therefrom, footnote, p. 320.

temperature, rate of evaporation, and load. It is also possible that steps 2 and 3 in this process may have a different order from that assumed. This is, however, of small consequence here. The principle to be recognized is that potential supersaturation will be reached in the liquid layer adjacent to the unloaded crystal first, and in the layer adjacent to the loaded crystal later, if at all. If no supersaturation occurred, obviously no growth of the loaded crystal would be possible. Growth at the bottom of the loaded crystal may therefore be positive, zero, or negative, and is much more likely to be zero or negative than positive in the conditions described by Bruhns and Mecklenburg. The rate of evaporation and the amount of the load are governing conditions here, supposing always that no other nuclei develop.

Two sets of measurements are submitted in support of this analysis. Table IV contains the record of two crystals, a loaded (74 gm.) and an unloaded one, placed in a saturated solution together, after the manner of Bruhns and Mecklenburg, and the results confirm their observations (Table II) perfectly. The unloaded crystal grows at the top, sides, and in the supporting rim below, while the loaded crystal shows no growth in the direction of the load.

TABLE IV  
CONDITIONS AS BEFORE

TIME (HOURS)	CRYSTAL 1 (LOAD 74 GM.)		CRYSTAL 2 (UNLOADED)	
	Thickness of Crystal	Increase of Thickness	Thickness of Crystal	Increase of Thickness
0.....	4.5988 mm.	0 mm.	3.9852 mm.	0 mm.
20.....	4.5988	-0.003	4.2588	0.274
43.....	4.6001	-0.001	4.6636	0.678

The results of Table V may perhaps serve to illustrate wherein Bruhns and Mecklenburg were hasty in generalizing from such an observation to the sweeping conclusion that no growth can take place in a loaded crystal in the direction in which the load is applied. This experiment (Table V) also shows two crystals, a loaded and an unloaded one, in the same saturated solution, and differs in no detail from the previous case (Table IV) save that the load has been

lightened (74 gm. to 0.7 gm.) and differs in no detail from Bruhns and Mecklenburg's series (Table II) save that our rate of evaporation from the solution was probably faster than theirs. The measurements leave nothing to be desired in the simplicity and directness of the proof offered that a loaded crystal may also lift its load in a solution containing an unloaded one (as unsuccessfully attempted by Bruhns and Mecklenburg), though in the light of the foregoing analysis it must be clear that the conditions there are least favorable for such growth. Nevertheless, growth will occur there also if the rate of evaporation is sufficiently high. It may not be inferred, however, that the supporting rim of the unloaded crystal carries no weight ("Er muss ja vornehmlich den Kristall

TABLE V  
CONDITIONS AS BEFORE

TIME (HOURS)	CRYSTAL 1 (LOAD 0.7 GM.)		CRYSTAL 2 (NO LOAD)	
	Thickness of Crystal	Increase of Thickness	Thickness of Crystal	Increase of Thickness
0.....	4.0307 mm.	0 mm.	3.4617 mm.	0 mm.
18.....	4.0357	0.005	3.5516	0.090
43.....	4.0492	0.019	3.9907	0.529

tragen," Bruhns and Mecklenburg, p. 105);<sup>†</sup> it merely carries less weight than the corresponding portion of the loaded crystal. The conditions in the liquid layers adjacent to the two crystals therefore differ in degree only, and one crystal may grow, or both may grow, according to the degree of supersaturation developed by the conditions of evaporation. Since the process of distribution of the crystalline molecules depends upon diffusion, a small difference of load means but a small difference in concentration at unit distance, and consequently a sluggish molecular flow. Diffusion would be similarly delayed by increasing the horizontal distance between a heavily weighted crystal and an unweighted one, but we have not put this evident inference to the test of experiment.

Had Bruhns and Mecklenburg chanced to place their unloaded crystal beneath the overhanging load upon its neighbor, it would

<sup>†</sup> See footnote, p. 320.

have grown alone until it shared the load, after which both crystals would have grown at rates approximately in inverse relation to their individual shares.

The effect of distributing a load over three crystals is shown by a simple case (Table VI).

TABLE VI  
EXPOSURE 3 DAYS. TEMPERATURE ABOUT 20°. LOAD 200 GM.

	THICKNESS OF CRYSTAL		INCREASE OF THICKNESS
	Initial	Final	
Crystal 1 . . . . .	3.416 mm.	3.527 mm.	0.111 mm.
Crystal 2 . . . . .	2.841	2.970	0.129
Crystal 3 . . . . .	3.033	3.108	0.075

Nor is it necessary that the measurements given in this table should stand alone in support of so important a conclusion. Bruhns and Mecklenburg succeeded in raising disks of porcelain loaded with weighted beaker-glasses, by the help of the crystallization of chrome alum, to elevations of a millimeter and more under certain conditions. These conditions were that the solution in contact with the disks and their load should be allowed to evaporate to dryness, the maximum elevations resulting when fresh portions of saturated solution were added and evaporated successively. These observations were not measured in detail, but are sufficiently well described in the following paragraphs (Bruhns and Mecklenburg, *op. cit.* pp., 107 and 108):

"Greater or less loading of the little beaker-glasses made no difference in the crystal development. All the six beakers—and this is the important point—no longer rested upon the porcelain disk but rather upon chrome alum crystals. After breaking the glasses loose, it was possible to establish beyond doubt that the crystals which supported the loaded beaker-glasses were at least 1 mm. thick, in many places even thicker, and the glass was no longer anywhere in contact with the porcelain supports" [p. 107].

"After a fragment of the crystallizing dish was broken away, in order to obtain a vertical section through the system, it could also be plainly seen that the porcelain disks no longer touched the bottom of the dish at any point, but were separated from it by a crystal layer the measured thickness of which was

from 1 to 2 mm. Through the crystallization of the chrome alum, therefore, both the porcelain support and the beaker-glasses were actually separated from the bottom and lifted up" [p. 108].

This elevation, they assert, has nothing to do with any force of crystallization, and is to be accounted for by "forces of capillarity and adsorption," but just how these forces produce this result they do not explain.

If two parallel plane surfaces are separated by a drop of liquid which wets them both, it is well known that the effect of capillarity is to urge them together with a force  $2TV/d^2$ , where  $T$  is the surface tension,  $V$  the volume of the drop, and  $d$  the distance between the plates, and this force may be great enough to change the melting-point of blocks of ice or to break sheets of heavy plate glass. Thus a flat disk of porcelain moistened with alum solution and resting on a flat glass plate experiences a downward pressure as if it were heavily loaded.

If a solution has a greater or smaller surface tension than the solvent, the solute tends respectively to desert the surface of the solution, or to seek it, and in the physics of colloids the concentration of solute at such a surface is known by analogy as adsorption. As E. Dorsey<sup>1</sup> and others have shown, aqueous solutions of salts such as chlorides and carbonates of the alkalis and zinc sulphate have a surface tension greater than that of water, and according to Poynting and Thomson,<sup>2</sup> the surface tension of salt solutions generally exceeds that of water. The adsorption in this case, considered as a possible lifting force, is therefore negative. Adsorption films in their relation to crystal growth have been investigated experimentally with great thoroughness by Marc in a series of four papers, "On Crystallization from Aqueous Solutions" (1908-10);<sup>3</sup> and

<sup>1</sup> *Phil. Mag.*, XLIV (1897), 369.

<sup>2</sup> *Properties of Matter*, 1902, p. 181.

<sup>3</sup> The chief conclusions reached by Marc are contained in the following brief citations (*Zeitschr. f. phys. Chem.*):

"It was found that the rate of crystallization, so far as it could be determined, of all the substances investigated was proportional to the square of the supersaturation" (*op. cit.*, LXVII [1909], 500).

"... that a very rapid change precedes the crystallization proper, which is interpreted to be an adsorption phenomenon. Support is given to this view by the

again, "On Adsorption and Saturated Surfaces" (1913),<sup>1</sup> without developing any single fact in support of the hypothesis advanced by Bruhns and Mecklenburg.

Neither capillarity nor adsorption exerts any upward pressure on the loaded disks of porcelain in the experiments under discussion, while adsorption does not prevent the exercise of the very great downward pressure due to the surface tension of water. Yet the alum crystallized and the disks were raised.

In the opinion of the observers it was essential to the elevation of the disks that evaporation should be complete.<sup>1</sup> Was the elevation, then, produced after the crystallization was complete and the mass solidified? The observers make no such statement, which, indeed, would seem absurd. But if the raising was not effected after solidification, it must have been produced before solidification, or while the underlying film was liquid and while crystallization was in progress, in opposition to capillary force as well as to the weight of the disks and their load.

Liesegang appears to have appreciated this anomaly in Bruhns and Mecklenburg's statement, though accepting their conclusion, for he sought to relieve it by the following explanation (referring to the experiments of Bruhns and Mecklenburg): "Nicht ein Wachstumsdruck der Krystalle sondern Capillar- und Adsorptionskräfte bewirkten hier also die Hebung. Das heisst die Leistung war schon vollbracht ehe Kristalle auftraten."<sup>2</sup> "The lifting was done before the crystals formed." This is not claimed by Bruhns and Mecklenburg, nor supported by any experimental evidence which they

fact that this preliminary phenomenon is particularly sensitive to slight impurities upon the crystal surface" (*ibid.*, LXXIII [1910], 718).

"No relation could be established between concentration and the quantity of adsorbed material" (*ibid.*, LXXIII [1910], 686).

"In all cases the rate of crystallization is diminished by the addition of substances which are adsorbed by the crystal, eventually even to the point of becoming practically zero" (*ibid.*, LXXIII [1910], 718).

"It was shown that the substances chiefly adsorbed by crystals are colloids, while the crystalloids are adsorbed only very slightly" (*ibid.*, LXXXI [1913], 692).

<sup>1</sup> Bruhns and Mecklenburg, *op. cit.*, p. 106: "Es sei aber ausdrücklich betont dass der Versuch nicht gelang, wenn wir nicht die Masse bis zum Grunde trocken werden liessen."

<sup>2</sup> R. E. Liesegang, "Kristallisationskraft," *Naturw. Rundschau der Chem. Ztg.*, Zweite Jahrg. 1913, p. 183.



offer, and indeed would seem to be without any foundation whatsoever.

We fail to see any reason for connecting the rise of the porcelain disks with capillarity or with adsorption. These could only obstruct the elevation, and must have been overcome by a linear force attending the crystallization of the alum, as in our own experiments.

It is not expedient, however, to rely on reasoning alone in matters of physics if experimentation is practicable, and we accordingly made the effort to separate the forces to which Bruhns and Mecklenburg appeal, through evaporation of solution of a colloid (gum arabic) in which was immersed a block of glass replacing the alum crystal between the two plates of glass (Table VII). Evaporation to dryness caused no rise of the upper glass plate as it

TABLE VII

BLOCK OF GLASS REPLACING THE ALUM CRYSTAL (FIG. 1). LOAD (GLASS PLATE) = 24 GM. GLASS BLOCK AND LOAD COMPLETELY IMMERSED IN 2 PER CENT SOLUTION OF GUM ARABIC IN WATER. ROOM TEMPERATURE

Time (Hours)	Spherometer Readings	Increase in Thickness	Notes
0.....	37.2609	0 mm.	
3.1.....	37.2649	+0.004	
28.1.....	37.2654	+0.0045	
45.1.....	37.2621	+0.001	
50.2.....	37.2615	+0.001	Evaporated to dryness
Refilled with 2 per cent gum arabic solution; all conditions unchanged			
58.8.....	37.2669	+0.006	
68.8.....	37.2670	+0.006	
76.1.....	37.2670	+0.006	
116.9.....	37.2653	+0.004	Evaporated to dryness

should have done were capillarity and adsorption the source of energy. A saturated solution of alum added to the colloid (Table VIII) starts crystal formation and growth at once, but at a rate much slower than in the cases where no colloid was present. This is in full accord with the experiments of Marc.<sup>1</sup>

In addition to confirming the results of Marc, Table VIII offers independent and explicit experimental proof that the "linear force" appears here also in spite of the action of the colloid in retarding

<sup>1</sup> See footnote, p. 326.

diffusion through increased viscosity and in interposing an adsorption film at the crystal surface.

Conditions in ore deposits appear to correspond very well with those in the laboratory, for crystallization may be found accompanied by local evidence of linear thrust or not, according to the magnitude and distribution of the opposing forces. Its failure is most often manifest in comb structure, found in crevices whose walls are each lined with tightly adhering crystals which either interlock and extend quite across the crevice or grow together near the central plane and mutually exclude further development. Such comb structure is common in veins, but far from universal.

TABLE VIII

SAME PLATES, BLOCK OF GLASS, AND CONDITIONS, EXCEPT THAT THE 2 PER CENT GUM ARABIC SOLUTION HAS BEEN SATURATED WITH POTASSIUM ALUMINIUM SULPHATE

Time (Hours)	Spherometer Readings	Increase in Thickness
0 .....	37.2653	0 mm.
4.2 .....	37.2694	0.004
24 .....	37.2721	0.007
29 .....	37.2812	0.016
46 .....	37.2818	0.017
71 .....	37.2842	0.019
95 .....	37.2857	0.020
148 .....	37.2873	0.022
173 .....	37.2916	0.026
214 .....	37.3079	0.043
287 .....	37.3178	0.053
366 .....	37.3170	0.052
409 .....	37.3185	0.053

It may be inferred, further, that linear pressure plays a subordinate part in much more complex occurrences.

Messrs. Bruhns and Mecklenburg seem to have misunderstood the last paragraph of our paper in which we called attention to the fact that the linear force of growing crystals *cannot* be disposed of as a mystery comparable to the growth of plant roots. It is a sharply defined physical process open to quantitative experimental investigation. It may not be fully understood, but it is no mystery.

The conclusion of these authors seems to be that during growth, material is added only to the upper and lateral faces of the crystal, so that a molecule once added remains at its original level. This was Kopp's contention in opposition to Lavalle, whose conclusions, however, were confirmed by Lehmann and others, including ourselves. This is in fact the root of the matter. If a given increment of the mass after deposition remains at its original level throughout the subsequent growth of the crystal, this exerts no linear force; while if the motion of the particle has a vertical component in consequence of the vertical extension of the lateral faces of the crystal, linear force is exerted.

On the other hand, if several crystals are immersed, one or more of them being loaded while others are not loaded, the loaded crystals grow only when the concentration of the solution in contact with them exceeds the saturation concentration for each crystal. Pressure, of course, increases solubility or raises the point of saturation for most salts.<sup>1</sup> Hence in such circumstances the unloaded crystals, or, more strictly, the less loaded crystals, usually are the only ones to exert lifting power, but in this case, also, growth raises the weight of each crystal.

Thus Bruhns and Mecklenburg's results with loaded porcelain disks are readily explicable. They experimented with solutions containing many small crystals, some of them weighted, others free. The disks did not rise measurably until the liquid was low and its surface (and consequent rate of evaporation) greatly increased by protruding solid matter, or until the crystals reached from the bottom of the dish to the disks, after which the disks were lifted.

Repetitions of this operation, extending over a few days, produced aggregate displacements of 5.0 mm. If to this be added our original measurement, twice confirmed in the course of the present control tests, that this linear force, because of the narrow rim

<sup>1</sup> As is well known, if the solution of a solid at constant temperature is attended by a diminution in total volume and a liberation of heat, pressure increases solubility. Such is the case for most crystalline solids including the alums. If the change in volume accompanying solutions is an increase, as in ammonium chloride, pressure decreases solubility.

through which it acts, actually exerts a pressure of the same order of magnitude as the breaking load of the solid crystals, need there be further hesitation in assuming that this is a force to be reckoned with in engineering<sup>1</sup> or in geology?<sup>2</sup>

## SUMMARY

In 1905 we showed by appropriate experimental evidence that a single crystal immersed in its own saturated solution, and growing by reason of the potential supersaturation of the solution resulting from evaporation will lift a weight placed upon it. This observation has been confirmed in the present paper.

In 1913 Bruhns and Mecklenburg placed two crystals in a similar saturated solution, one loaded and the other free, and noted that the load upon the one crystal was not raised, although the free crystal grew rapidly. From this experiment they were led to deny the power of a crystal to lift a weight of foreign substance, although admitting the power of the unloaded crystal to lift its own substance. They appear to have overlooked in this conclusion the fact that the solubility of the loaded crystal is for most substances greater than that of an unloaded one, and also that this is a difference in degree only, for the unloaded crystal also supports weight (its own).

In consequence of this greater solubility, with an unloaded and a loaded crystal in the same solution, the necessary condition of potential supersaturation will be reached in the liquid adjacent to the unloaded crystal before it is reached in the other, and the growth of the unloaded crystal thereafter may keep the concentration below that necessary for the growth of the loaded crystal. This appears to be the condition reached in Bruhns and Mecklenburg's experiment. If it happens, however, that the rate of growth

<sup>1</sup> Cf. the investigations of Dr. Hans Kühle, "Die Ursache des Treibens der Zemente," *Tonindustrie Ztg.*, XXXVI (1912), 1331-34; and of Klein and Phillips, "Hydration of Portland Cement," *Technologic Papers of the Bureau of Standards* No. 43 (1914), pp. 50, 56, 57.

<sup>2</sup> Cf. the recent observations of Stephen Taber, *Virginia Geol. Survey Bull.*, No. VII (1913), p. 222; also G. D. Harris, "Rock Salt, Its Origin, Geological Occurrences and Economic Importance in the State of Louisiana," *Geol. Survey of Louisiana, Bulletin* No. 7 (1907), p. 75.

of the unloaded crystal is insufficient to take up all of the excess concentration provided by the continued evaporation, then supersaturation will increase. It is entirely possible under these conditions that the potential supersaturation necessary for the growth of the loaded crystal may then be attained or even exceeded, and that the loaded crystal will also grow and lift its load. This condition was attained experimentally without difficulty in the observations recorded in this paper. If concentration increases still more rapidly, and exceeds the ability of both unloaded and loaded crystals to take up, through their continued growth, all the matter in excess of the saturation concentration, then additional nuclei may form upon which excess matter may be deposited. This appears to have been the condition attained in the last series of Bruhns and Mecklenburg's observations in which the solution was evaporated to dryness.

Here six disks of porcelain loaded with weights were all raised a millimeter or more in the same solution, but Bruhns and Mecklenburg attribute this result to the action of capillarity and adsorption, and deny the competence of the "linear force of growing crystals" to effect such mechanical displacements.

A simple analysis suffices to show that capillarity in a solution evaporating to dryness can have no other effect than to press the crystal down upon its base with a force equal to  $2TV/d^2$ , where  $T$  is the surface tension,  $V$  the volume of the drop of liquid between the crystal and its base, and  $d$  the distance separating the two, and that the lifting action observed by Bruhns and Mecklenburg has occurred in spite of this opposing force and not because of it. Adsorption delays diffusion and diminishes the rate of growth, but does nothing to promote it. These forces therefore cannot be appealed to in explanation of the lifting observed by Bruhns and Mecklenburg and by us.

We therefore return to the original thesis that the growth of crystals in saturated solution develops a linear force in the direction of the load, and that neither the magnitude of the load (up to the breaking load) nor its character (whether exclusively crystal substance or partly foreign substance) has any other effect than to

increase solubility and so to raise the concentration necessary for potential supersaturation and growth upon the loaded crystals. This degree of supersaturation is readily attainable through evaporation or otherwise, and when attained the loads are lifted. With this thesis established, there is no conflict between the observations of Bruhns and Mecklenburg and our own, and all the experimental evidence offered is perfectly correlated.

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